Docket No.: 14113-00050-US
(PATENT)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:
Ingrid Bach et al.

Application No.: 10/559,749 | Confirmation No.: 5947

Filed: January 19, 2006 | Art Unit: 1626

For: METHOD FOR PRODUCING HIGHLY PURIFIED, TRIS- AND BIS-ORTHO-METALLATED ORGANOMETALLIC COMPOUNDS | Examiner: J. R. Kosack

1.132 Declaration

MS Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

1. I, Dr. Philipp Stoessel am a citizen of the Federal Republic of Germany and reside at Sophienstrasse 30, 60487 Frankfurt, Germany, hereby declare and say as follows:

2. I am a fully trained chemist, having studied chemistry at the University of Tuebingen, Baden-Wuerttemberg, Germany.
I am well acquainted with technical English.

3. Work experience:
1986 – 1996: Studies in chemistry at the University of Tuebingen, Germany
1996 – 1998: Postdoctoral research in chemistry sponsored by the Alexander von Homboldt Foundation with Dr. J. R. Norton at the Colorado State University, Fort Collins, CO and at the Columbia University New York, NY.
1998 – 1999: Material Scientist and Synthetic Chemist at the Institut fuer Neue Materialien, Saarbruecken, Saarland, Germany

1999 – 2005: Material Scientist and Synthetic Chemist at COVION Organic Semiconductors GmbH, Frankfurt, Hessen, Germany

2005 – 2007: Material Scientist and Synthetic Chemist at the Merck Organic Materials GmbH, Frankfurt, Hessen, Germany

2007 – today: Material Scientist and Synthetic Chemist at the Merck KGaA, Frankfurt, Hessen, Germany

4. In the field of organic light emitting diode (OLED), I am an inventor on more than 75 U.S. patents and patent applications and an author of more than 10 publications and lectures.

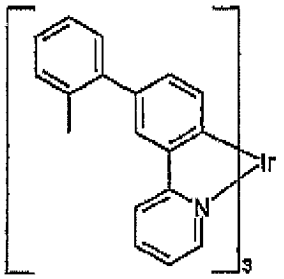
5. In view of my qualifications as outlined above, I consider myself to be an expert and to be skilled in the metal complex used in the electroluminescent devices such as organic light emitting diodes ("OLED").

6. I am one of the inventors in Application No.: 10/559,749 ("749 application) which published as U.S.20060142552 (published application).

7. Under my supervision I had the following experiments based on the disclosure the '749 application.

8. a) ***fac-Tris[2-(2-pyridinyl-κN)(4-(2-methylphenyl)phenyl-κC]iridium(III)***

Inventive example

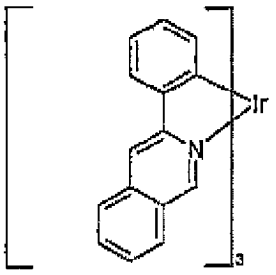


0.484 g (1.0 mmol) of $Na[IrCl_2(acac)_2]$ and 2.45 g (10 mmol) of 2-(2'-methyl[1,1'-biphenyl]-3-yl)pyridine [875462-70-7] are added to 10 ml of degassed ethylene glycol.

The suspension is exposed at 190°C to a microwave radiation as specified in paragraph [0058] of the publication application with good stirring for 15 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a yellow, finely crystalline precipitate is poured with stirring into a mixture of 20 ml of aqueous 1 N hydrochloric acid and 60 ml of ethanol. After stirring for 5 minutes, the mixture is filtered with suction through a glass suction filter (P3), and the yellow, finely crystalline precipitate is washed three times with 5 ml each time of aqueous 1 N hydrochloric acid and five times with 5 ml each time of water and five times with 5 ml each time of ethanol, and subsequently dried under high vacuum at 80°C for 5 h and at 200°C for 2 h. The yield, at a purity of > 99.9% by HPLC, is 0.856 g, corresponding to 92.5 %.

b) fac-Tris[2-(3-isoquinolinyl-κN)phenyl-κC]iridium(III)

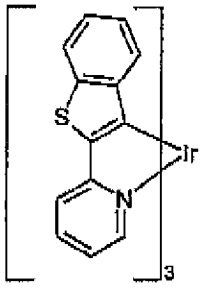
Inventive example



0.484 g (1.0 mmol) of $Na[IrCl_2(acac)_2]$ and 2.05 g (10 mmol) of 3-phenylisoquinoline [37993-76-3] are added to 10 ml of degassed ethylene glycol. The suspension is exposed at 170°C to an above-specified microwave radiation with good stirring for 15 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a deep red precipitate is poured with stirring into a mixture of 20 ml of aqueous 1 N hydrochloric acid and 60 ml of ethanol. After stirring for 5 minutes, the mixture is filtered with suction through a glass suction filter (P3), and the deep red precipitate is washed three times with 5 ml each time of aqueous 1 N hydrochloric acid and five times with 5 ml each time of water and five times with 5 ml each time of ethanol, and subsequently dried under high vacuum at 80°C for 5 h and at 200°C for 2 h. The yield, at a purity of > 99.7% by HPLC, is 0.720 g, corresponding to 89.4 %.

c) fac-Tris[2-(2-pyridinyl-κN)benzo[b]thien-3-yl-κC]iridium(III)

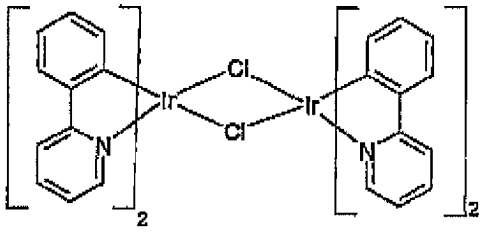
Inventive example



0.484 g (1.0 mmol) of $Na[IrCl_2(acac)_2]$ and 2.11 g (10 mmol) of 2-benzo[b]thien-2-yl-pyridine [38210-35-4] are added to 10 ml of degassed ethylene glycol. The suspension is exposed at 150°C to an above-specified microwave radiation with good stirring for 25 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a reddish orange precipitate is poured with stirring into a mixture of 20 ml of aqueous 1 N hydrochloric acid and 60 ml of ethanol. After stirring for 5 minutes, the mixture is filtered with suction through a glass suction filter (P3), and the reddish orange precipitate is washed three times with 5 ml each time of aqueous 1 N hydrochloric acid and five times with 5 ml each time of water and five times with 5 ml each time of ethanol, and subsequently dried under high vacuum at 80°C for 5 h and at 200°C for 2 h. The yield, at a purity of > 99.9% by HPLC, is 0.731 g, corresponding to 88.8 %.

d) Di-μ-chloro-tetrakis[2-(2-pyridinyl-κN)phenyl-κC]di-iridium(III)

Inventive example

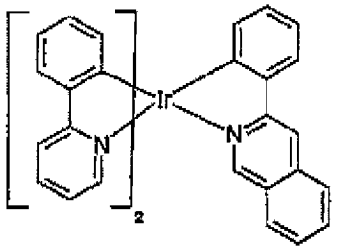


0.484 g (1.0 mmol) of $Na[IrCl_2(acac)_2]$ and 0.31 g (2.0 mmol) of 2-phenylpyridine are added to 10 ml of degassed ethylene glycol. The suspension is exposed at 150°C to an

above-specified microwave radiation with good stirring for 15 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a yellow, finely crystalline precipitate is poured with stirring into 50 ml of ethanol. After stirring for 30 minutes, the mixture is filtered with suction through a glass suction filter (P3), and the yellow, finely crystalline precipitate is washed five times with 5 ml each of ethanol, and subsequently dried under high vacuum at 80°C for 5 h. The yield, at a purity of > 99.8% by HPLC, is 0.501 g, corresponding to 93.4 %.

e) Bis[2-(2-pyridinyl-κN)phenyl-κC] [2-(3-isoquinolinyl-κN)phenyl-κC]iridium(III)

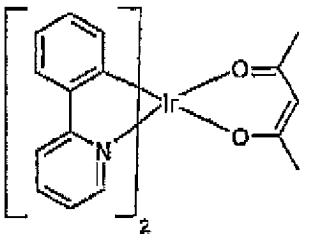
Inventive example



1.07 g (1.0 mmol) of di-μ-chloro-tetrakis[2-(2-pyridinyl-κN)phenyl-κC]di-iridium(III) and 0.431 g (2.1 mmol) of 3-phenylisoquinoline [37993-76-3] are added to 20 ml of degassed ethylene glycol. The suspension is exposed at 150°C to an above-specified microwave radiation with good stirring for 15 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a red, finely crystalline precipitate is poured with stirring into a mixture of 20 ml of aqueous 1 N hydrochloric acid and 100 ml of ethanol. After stirring for 5 minutes, the mixture is filtered with suction through a glass suction filter (P3), and the red, finely crystalline precipitate is washed three times with 5 ml each time of aqueous 1 N hydrochloric acid and five times with 5 ml each time of water and five times with 5 ml each time of ethanol, and subsequently dried under high vacuum at 80°C for 5 h and at 200°C for 2 h. The yield, at a purity of > 99.8% by HPLC, is 1.28 g, corresponding to 90.8 %.

f) Acetylacetonato-bis-[2-(2-pyridinyl-κN)phenyl-κC]iridium(III)

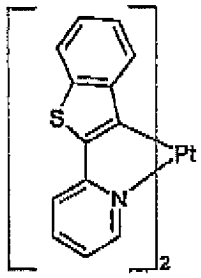
Inventive example



1.07 g (1.0 mmol) of di-μ-chloro-tetrakis[2-(2-pyridinyl-κN)phenyl-κC]di-iridium(III) and 0.62 ml (6.0 mmol) of acetylacetone are added to a degassed mixture of 10 ml of 2-ethoxy ethanol and 3 ml of water. The suspension is exposed at 60°C to an above-specified microwave radiation with good stirring for 10 min. After cooling to room temperature, the reaction mixture is filtered with suction through a glass suction filter (P3), and the yellow, finely crystalline precipitate is washed five times with 5 ml each of a mixture of ethanol and water (1:1, vv), and subsequently dried under high vacuum at 80°C for 5 h. The yield, at a purity of > 99.8% by HPLC, is 1.12 g, corresponding to 93.5%.

g) Bis[2-(2-pyridinyl-κN)benzo[b]thien-3-yl-κC]platinum(II)

Inventive example



0.472 g (1.0 mmol) of bis-benzonitrile-platinum(II)chloride and 1.27 g (6 mmol) of 2-benzo[b]thien-2-yl-pyridine [38210-35-4] are added to 10 ml of degassed ethylene glycol. The suspension is exposed at 110°C to an above-specified microwave radiation with good stirring for 15 min. After cooling to room temperature, the reaction mixture which contains the product in the form of a orange precipitate is diluted with 50 ml of ethanol. After stirring for 5 minutes, the mixture is filtered with suction through a glass suction filter (P4), and the orange precipitate is washed five times with 5 ml each time of

ethanol, and subsequently dried under high vacuum at 80°C for 5 h and at 150°C for 2 h. The yield, at a purity of > 99.5% by HPLC, is 0.288 g, corresponding to 46.6 %.

9. Examples a) – g) above, demonstrate the following:

a) reaction using a substituted phenylpyridine ligand to form a homoleptic iridium complex of formula (1), thus showing that the reaction does not only work with unsubstituted ligands, but also with substituted ligands;

b) reaction using a phenyl-isoquinoline ligand to form a homoleptic iridium complex of formula (1), thus showing that the reaction does not only work with pyridine ligands, but also with other N-heteroaromatic groups;

c) reaction using a benzothienyl-pyridine ligand to form a homoleptic iridium complex of formula (1), thus showing that the reaction does not only work with phenyl ligands, but also with other groups CyC;

d) reaction using a phenylpyridine ligand to form a chloro-bridged dimeric iridium complex of formula (5a), thus showing that the reaction does not only work for mononuclear complexes but also for dinuclear complexes;

e) reaction using a phenyl-isoquinoline ligand to form a heteroleptic iridium complex of formula (1), thus showing that the reaction does not only work for the synthesis of homoleptic complexes, but also for the synthesis of heteroleptic complexes;

f) reaction using an acetylacetonate ligand to form a heteroleptic iridium complex of formula (3), thus showing that the reaction does not only work for ortho-metalated ligands, but also for other chelating ligands X; and

g) reaction using a benzothienyl-pyridine ligand to form a homoleptic platinum complex of formula (1), thus showing that the reaction does not only work with iridium as metal M, but also with platinum.

10. The above examples establish that complexes of the formulae (1), (3) and (5a) can be produced according to the '749 specification.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Philipp Stoessel

11-16-09
Date